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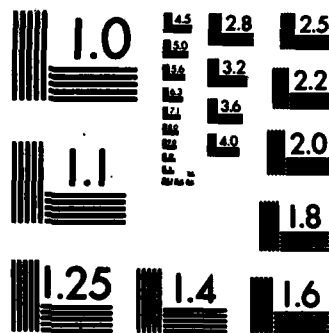
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A graph-theoretical approach is adopted for the complete systematization and enumeration of all the possible polytertiary phosphine structures of various types. The three differing types of structural variations in the polytertiary phosphines and related compounds involve (i) substitution of one or more of the phosphorus atoms by other group V atoms; (ii) the presence of two or more types of bridging groups; and (iii) the presence of two or more differing terminal groups. In determining appropriate cycle indices for each of these structural types, the number of isomers for each is automatically generated.

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Isomer Enumeration in Polytertiary Phosphines and

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by

R.B. King and D.H. Rouvray

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University of Georgia
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ABSTRACT

A graph-theoretical approach is adopted for the complete systematization and enumeration of all the possible polytertiary phosphine structures of various types. The three differing types of structural variations in the polytertiary phosphines and related compounds involve (i) substitution of one or more of the phosphorus atoms by other group V atoms; (ii) the presence of two or more types of bridging groups; and (iii) the presence of two or more differing terminal groups. In determining appropriate cycle indices for each of these structural types, the number of isomers for each is automatically generated.

Introduction

Over the past two decades synthetic methods have been developed for the preparation of polytertiary phosphines and arsines having a wide variety of structures¹⁻⁶. Several years ago an attempt was made to systematize the different types of polytertiary phosphines known at the time. The methods involved were ones which had proved effective for analysis of the mathematically similar, but chemically completely different, planar networks of sp^2 carbon atoms.^{7,8} In the present paper we extend this earlier work by presenting complete and systematic guidelines for the enumeration of the possible polytertiary phosphine structures of various types. Our approach is based on the mathematical discipline of graph theory.

The use of graph theory in the solution of chemical problems, especially those arising in isomer enumeration studies, is now generally well established. For further details of the applications of graph theory in the chemical context, the interested reader is referred to reviews on the subject by one of the present authors⁹⁻¹¹. In this new treatment we shall represent the framework of a given polytertiary phosphine by a spanning tree graph. The vertices of this graph correspond to the phosphorus atoms and the edges to the bridges between these atoms. In the actual enumeration use is made of the Enumeration Theorem of Pólya¹¹⁻¹³.

We shall consider here three differing types of structural variation in the polytertiary phosphines and related compounds. These three types comprise structures for which (i) one or more of the phosphorus atoms is substituted with other group V atoms such as nitrogen, arsenic, or antimony; (ii) two or more differing types of bridging group are present. Types of bridging groups frequently found in the polytertiary phosphines include the $-CH_2CH_2-$; cis and trans $=CH=CH-$; $-C=C-$; and o-, m-, and p-phenylene groups; (iii) two or more

differing types of terminal group are present. Terminal groups frequently found in the polytertiary phosphines include the hydrogen¹⁴, methyl³, neopentyl¹⁵, phenyl¹, dimethylamino¹⁶, methoxy¹⁶, and neomenthyl¹⁷ groups.

Representation of Polytertiary Phosphines

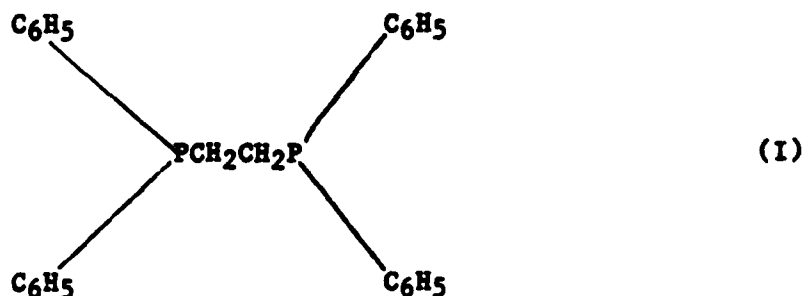
In general terms, a polytertiary phosphine may be regarded as a chemical species consisting of p phosphorus atoms linked together by b bridges, and containing a total of t terminal groups. From this general definition, two relationships may be derived which relate the three parameters p , b , and t . The trivalency of all of the phosphorus atoms in polytertiary phosphines implies that the number of bridges and terminal groups bonded to each of the p phosphorus atoms must always total three, this fact leading directly to the relationship:

$$3p = 2b + t \quad (1)$$

The representation of polytertiary phosphines as spanning trees with the vertices corresponding to the phosphorus atoms and the edges to the bridges yields the second relationship. Since the cyclomatic number¹⁸ of any tree graph is zero, the following relationship, must hold:

$$p - b = 1 \quad (2)$$

The above definitions may be illustrated by reference to the well-known ditertiary phosphine I:



This molecule has two phosphorus atoms ($p = 2$), one $-\text{CH}_2\text{CH}_2-$ bridge ($b = 1$),

and four terminal phenyl groups ($t = 4$). These numbers, it may be noted, satisfy our equations (1) and (2) above.

In Figure 1 we depict all the possible trees representing polytertiary phosphines containing up to six phosphorus atoms. It is of interest that these trees are identical to those representing acyclic networks of planar sp^2 carbon atoms.^{8,19} Interestingly, our trees are also identical to the so-called "boron-trees" first discussed by Cayley over a century ago²⁰.

The Enumeration Procedure

We start by determining the appropriate cycle indices for the three different structural types listed above. For polytertiary phosphine systems showing structural variation of type (i), i.e. structures in which phosphorus atoms are substituted by other group V atoms, the relevant cycle index is simply that for the permutations of the vertices of the corresponding tree. The trees T are those depicted in Figure 1, and their cycle indices we shall designate as $Z(T)$. In cases where the atoms at each vertex of the tree can be either one of two possible group V atoms, e.g. either phosphorus or arsenic, the coefficient of x^m in the expansion of the counting polynomial obtained from the cycle index $Z(T; 1+x)$ will give the number of isomeric molecules containing m atoms of the group V element. The appropriate cycle indices are listed in Table 1 and the corresponding counting polynomials are presented in Table 2.

In order to determine the number of isomers in polytertiary phosphine systems showing structural variation of type (ii), i.e. for the case, when differing bridging groups are present in the structure, it is first necessary to set up the cycle index $Z(G)$ of a graph G derived from the tree T . The graph G is constructed from T by the following procedure:

- (a) all vertices in T of degree 1 vanish in G ;
- (b) all edges in T become vertices in G ;

- (c) all vertices in T of degree 2 become edges in G connecting those vertices in G which correspond to the edges in T meeting at the vertex of degree 2;
- (d) all vertices in T of degree 3 become triangles in G . The vertices of such triangles correspond to those edges in T which meet at a vertex of degree 3.

The graphs G derived in this way from the trees T of Figure 1 are depicted in Figure 2. The cycle indices $Z(G)$ for these graphs G are listed in Table 1. In cases where two different bridges are present in a polytertiary phosphine network, e.g. $-\text{CH}_2\text{CH}_2-$ and cis- $\text{CH}=\text{CH}-$ bridges, the coefficient of \underline{x}^m in the counting polynomial obtained by the expansion of the cycle index $Z(G; 1+\underline{x})$ will indicate the number of isomers containing \underline{m} bridges of one of the two possible types. These counting polynomials are also to be found in Table 2.

To calculate the number of isomers for a given polytertiary phosphine network containing structural variations of type (iii), i.e. for structures containing two or more differing types of terminal group, it is necessary to determine the cycle index of the permutations of the vertices of degree 1 of an expanded tree T' derived from T by adding new edges to each vertex of T of degrees one or two until all the vertices originally belonging to T have degree three. The expanded trees T' constructed in this way are illustrated in Figure 3. The cycle indices $Z(T')$ of these expanded trees T' are listed in Table 1. In those cases where two differing terminal groups, e.g. methyl and phenyl, are present in a polytertiary phosphine network, the coefficient of \underline{x}^m in the counting polynomial obtained by expansion of the cycle index $Z(T'; 1+\underline{x})$ will indicate the number of isomers containing \underline{m} terminal groups of one of the two possible types. The appropriate counting polynomials are also listed in Table 2.

Conclusion

The above calculations on polytertiary phosphine networks have been restricted to systems displaying three differing types of structural variation. There are clearly several other, more complex variations of these isomer enumeration problems that could be treated by a natural extension of our approach. Moreover, in cases where isomers having three, four or more differing group V atoms, bridge types, or terminal groups are to be enumerated, it is possible to obtain the respective counting polynomials derived by substituting the expressions $1+x+y$, $1+x+y+z$, and so on, into the appropriate cycle index $Z(T)$, $Z(G)$, or $Z(T')$. In cases where different combinations of group V atoms, bridges, and terminal groups are involved and two or three of these items are to be varied, the total isomer count C will be given as the product of the isomer counts:

$$C = C_T \times C_G \times C_{T'} \quad (3)$$

where C_T , C_G and $C_{T'}$ refer respectively to the counts for each of the types of structural variation (i) to (iii) discussed in this paper. It is anticipated that the determinations described herein may well have ramifications outside the confines of phosphorus chemistry. In fact, our enumerations could have a bearing on, and even direct relevance to, a wide variety of systems not discussed here.

Acknowledgment

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Captions to Figures

Figure 1 : Graphs in the form of trees T representing the possible networks for polytertiary phosphine systems containing up to six phosphorus atoms.

Figure 2 : The graphs G derived from the trees T of Figure 1 (see text).

Figure 3 : The expanded trees T' derived from the trees T of Figure 1 (see text).



2A



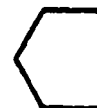
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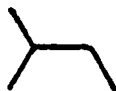
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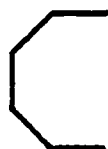
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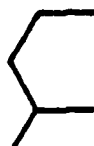
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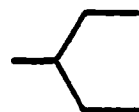
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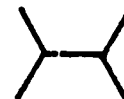
6A



6B



6C



6D



2A



3A



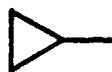
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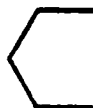
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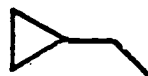
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5B



6A



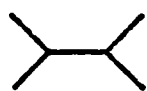
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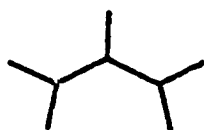
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6D



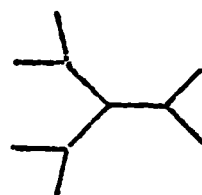
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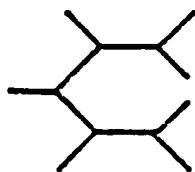
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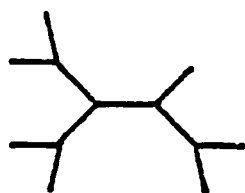
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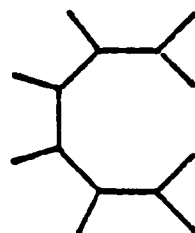
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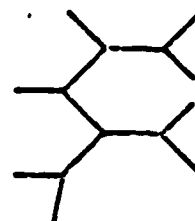
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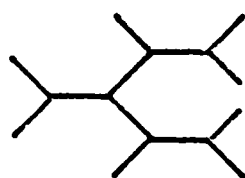
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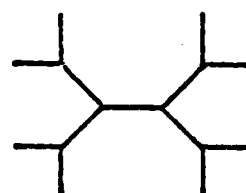
6A



6B



6C



6D

TABLE 1 : Cycle Indices for the Ten Possible Polytertiary Phosphine Networks with Two to Six Phosphorus Atoms

System	Cycle Index Z(T) for Phosphorus Atom Substitution	Cycle Index Z(G) for Bridge Substitution	Cycle Index Z(T') for Terminal Group Substitution
2A	$\frac{1}{2}(\bar{x}_1^2 + \bar{x}_2)$	\bar{x}_1	$\frac{1}{4}(\bar{x}_1^4 + 3\bar{x}_2^2)$
3A	$\frac{1}{2}(\bar{x}_1^3 + \bar{x}_1\bar{x}_2)$	$\frac{1}{2}(\bar{x}_1^2 + \bar{x}_2)$	$\frac{1}{4}(\bar{x}_1^5 + 3\bar{x}_1\bar{x}_2^2)$
4A	$\frac{1}{2}(\bar{x}_1^4 + \bar{x}_2^2)$	$\frac{1}{2}(\bar{x}_1^3 + \bar{x}_1\bar{x}_2)$	$\frac{1}{4}(\bar{x}_1^6 + \bar{x}_1^2\bar{x}_2^2 + 2\bar{x}_2^3)$
4B	$\frac{1}{6}(\bar{x}_1^4 + 2\bar{x}_1\bar{x}_3 + 3\bar{x}_1^2\bar{x}_2)$	$\frac{1}{6}(\bar{x}_1^3 + 2\bar{x}_3 + 3\bar{x}_1\bar{x}_2)$	$\frac{1}{6}(\bar{x}_1^6 + 2\bar{x}_3^2 + 3\bar{x}_2^3)$
5A	$\frac{1}{2}(\bar{x}_1^5 + \bar{x}_1\bar{x}_2^2)$	$\frac{1}{2}(\bar{x}_1^4 + \bar{x}_2^2)$	$\frac{1}{4}(\bar{x}_1^7 + 2\bar{x}_1\bar{x}_2^3 + \bar{x}_1^3\bar{x}_2^2)$
5B	$\frac{1}{2}(\bar{x}_1^5 + \bar{x}_1^3\bar{x}_2)$	$\frac{1}{2}(\bar{x}_1^4 + \bar{x}_1\bar{x}_2)$	$\frac{1}{2}(\bar{x}_1^7 + \bar{x}_1\bar{x}_2^3)$
6A	$\frac{1}{2}(\bar{x}_1^6 + \bar{x}_2^3)$	$\frac{1}{2}(\bar{x}_1^5 + \bar{x}_1\bar{x}_2^2)$	$\frac{1}{4}(\bar{x}_1^8 + \bar{x}_1^4\bar{x}_2^2 + 2\bar{x}_2^4)$

TABLE 1 : Cycle Indices for the Ten Possible Polytertiary Phosphine Networks with Two to Six Phosphorus Atoms

System	Cycle Index Z(T) for Phosphorus Atom Substitution	Cycle Index Z(G) for Bridge Substitution	Cycle Index Z(T') for Terminal Group Substitution
6B	$\frac{1}{2}(\bar{x}_1^6 + \bar{x}_1^4 \bar{x}_2^1)$	$\frac{1}{2}(\bar{x}_1^5 + \bar{x}_1^3 \bar{x}_2^1)$	$\frac{1}{2}(\bar{x}_1^8 + \bar{x}_1^2 \bar{x}_2^3)$
6C	$\frac{1}{2}(\bar{x}_1^6 + \bar{x}_1^2 \bar{x}_2^2)$	$\frac{1}{2}(\bar{x}_1^5 + \bar{x}_1^1 \bar{x}_2^2)$	$\frac{1}{2}(\bar{x}_1^8 + \bar{x}_2^4)$
6D	$\frac{1}{4}(\bar{x}_1^6 + 2\bar{x}_2^3 + \bar{x}_1^2 \bar{x}_2^2)$	$\frac{1}{4}(\bar{x}_1^5 + 3\bar{x}_1^1 \bar{x}_2^2)$	$\frac{1}{4}(\bar{x}_1^8 + 3\bar{x}_2^4)$

TABLE 2 : Counting Polynomials for the Ten Possible Polytertiary Phosphine Networks with Two to Six Phosphorus Atoms

System	Polynomial $Z(T; 1+\bar{x})$ for Phosphorus Atom Substitution	Polynomial $Z(G; 1+\bar{x})$ for Bridge Substitution	Polynomial $Z(T'; 1+\bar{x})$ for Terminal Group Substitution
2A	$1 + \bar{x} + \bar{x}^2$	$1 + \bar{x}$	$1 + \bar{x} + 3\bar{x}^2 + \bar{x}^3 + \bar{x}^4$
3A	$1 + 2\bar{x} + 2\bar{x}^2 + \bar{x}^3$	$1 + \bar{x} + \bar{x}^2$	$1 + 2\bar{x} + 4\bar{x}^2 + 4\bar{x}^3 + 2\bar{x}^4 + \bar{x}^5$
4A	$1 + 2\bar{x} + 4\bar{x}^2 + 2\bar{x}^3 + \bar{x}^4$	$1 + 2\bar{x} + 2\bar{x}^2 + \bar{x}^3$	$1 + 2\bar{x} + 6\bar{x}^2 + 6\bar{x}^3 + 6\bar{x}^4 + 2\bar{x}^5 + \bar{x}^6$
4B	$1 + 2\bar{x} + 2\bar{x}^2 + 2\bar{x}^3 + \bar{x}^4$	$1 + \bar{x} + \bar{x}^2 + \bar{x}^3$	$1 + \bar{x} + 4\bar{x}^2 + 4\bar{x}^3 + 4\bar{x}^4 + \bar{x}^5 + \bar{x}^6$
5A	$1 + 3\bar{x} + 6\bar{x}^2 + 6\bar{x}^3 + 3\bar{x}^4 + \bar{x}^5$	$1 + 2\bar{x} + 4\bar{x}^2 + 2\bar{x}^3 + \bar{x}^4$	$1 + 3\bar{x} + 8\bar{x}^2 + 12\bar{x}^3 + 12\bar{x}^4 + 8\bar{x}^5 + 3\bar{x}^6 + \bar{x}^7$
5B	$1 + 4\bar{x} + 7\bar{x}^2 + 7\bar{x}^3 + 4\bar{x}^4 + \bar{x}^5$	$1 + 3\bar{x} + 4\bar{x}^2 + 3\bar{x}^3 + \bar{x}^4$	$1 + 4\bar{x} + 12\bar{x}^2 + 19\bar{x}^3 + 19\bar{x}^4 + 12\bar{x}^5 + 4\bar{x}^6 + \bar{x}^7$

TABLE 2 : Counting Polynomials for the Ten Possible Polytertiary Phosphine Networks with Two to Six Phosphorus Atoms

System	Polynomial $Z(T; 1+\bar{x})$ for Phosphorus Atom Substitution	Polynomial $Z(G; 1+\bar{x})$ for Bridge Substitution	Polynomial $Z(T'; 1+\bar{x})$ for Terminal Group Substitution
6A	$1 + 3\bar{x} + 9\bar{x}^2 + 10\bar{x}^3 + 9\bar{x}^4 + 3\bar{x}^5 + \bar{x}^6$	$1 + 3\bar{x} + 6\bar{x}^2 + 6\bar{x}^3 + 3\bar{x}^4 + \bar{x}^5$	$1 + 3\bar{x} + 11\bar{x}^2 + 17\bar{x}^3 + 24\bar{x}^4 + 17\bar{x}^5 + 11\bar{x}^6 + 3\bar{x}^7 + \bar{x}^8$
6B	$1 + 5\bar{x} + 11\bar{x}^2 + 14\bar{x}^3 + 11\bar{x}^4 + 5\bar{x}^5 + \bar{x}^6$	$1 + 4\bar{x} + 7\bar{x}^2 + 7\bar{x}^3 + 4\bar{x}^4 + \bar{x}^5$	$1 + 5\bar{x} + 16\bar{x}^2 + 31\bar{x}^3 + 38\bar{x}^4 + 31\bar{x}^5 + 16\bar{x}^6 + 5\bar{x}^7 + \bar{x}^8$
6C	$1 + 4\bar{x} + 9\bar{x}^2 + 12\bar{x}^3 + 9\bar{x}^4 + 4\bar{x}^5 + \bar{x}^6$	$1 + 3\bar{x} + 6\bar{x}^2 + 6\bar{x}^3 + 3\bar{x}^4 + \bar{x}^5$	$1 + 4\bar{x} + 16\bar{x}^2 + 28\bar{x}^3 + 38\bar{x}^4 + 28\bar{x}^5 + 16\bar{x}^6 + 4\bar{x}^7 + \bar{x}^8$
6D	$1 + 2\bar{x} + 6\bar{x}^2 + 6\bar{x}^3 + 6\bar{x}^4 + 2\bar{x}^5 + \bar{x}^6$	$1 + 2\bar{x} + 4\bar{x}^2 + 4\bar{x}^3 + 2\bar{x}^4 + \bar{x}^5$	$1 + 2\bar{x} + 10\bar{x}^2 + 14\bar{x}^3 + 22\bar{x}^4 + 14\bar{x}^5 + 10\bar{x}^6 + 2\bar{x}^7 + \bar{x}^8$

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